LA-UR- 01-4479

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Two energy scales and slow crossover in YbAl$_3$

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(Dated: July 31, 2001)

Experimental results for the susceptibility, specific heat, 4f occupation number, Hall effect and magnetoresistance for single crystals of YbAl$_3$ show that, in addition to the Kondo energy scale $k_B T_K \sim 670$K, there is a low temperature scale $T_{coh} < 50$K for the onset of coherence. Furthermore the crossover from the low temperature Fermi liquid regime to the high temperature local moment regime is slower than predicted by the Anderson impurity model. These effects may reflect the behavior of the Anderson Lattice in the limit of low conduction electron density.

The low temperature transport behavior of periodic intermediate valent (IV) and heavy fermion (HF) compounds[1] is fundamentally different from that expected for the Anderson impurity model (AIM), in that it manifests vanishing resistivity (Bloch's law) and an optical conductivity[2] appropriate for renormalized band behavior. Similarly, the 4f electrons have a coherent effect on the Fermi surface, as seen in de Haas van Alphen (dHvA) measurements[3]. However the temperature dependence of the susceptibility, specific heat and 4f occupation number and the energy dependence of the dynamic susceptibility show behavior that is qualitatively very similar to the predictions of the AIM[4, 5]. Essentially this is because these properties are dominated by spin/valence fluctuations that are highly local and which exhibit a Lorentzian power spectrum[6] consistent with the AIM.

Nevertheless, recent theoretical studies[7, 8] of the Anderson lattice (AL) in the Kondo limit suggest that these latter properties can differ in at least two ways from the predictions of the AIM. First, as the background conduction electron density n_c decreases, the theory predicts[8] a new low temperature scale T_{coh} for the onset of Fermi liquid coherence, where T_{coh} is significantly smaller than the high temperature Kondo scale T_K. For $T < T_{coh}$ peaks in addition to those expected on the high temperature scale T_K are predicted for the susceptibility and specific heat. Second, as n_c decreases the crossover from low temperature Fermi liquid behavior to high temperature local moment behavior becomes slower (i.e. more gradual) than predicted for the AIM[7].

Recently we have given evidence[5] for a slow crossover of the susceptibility and 4f occupation number in the IV compounds YbXCu$_4$ (X = Ag, Cd, Mg, Tl, Zn). In this paper we give evidence *both* for a slow crossover *and* for two energy scales in the IV compound YbAl$_3$. We show that both effects are observed not only for the susceptibility but also for the specific heat. We show that the magnetotransport gives clear evidence for a change in character on the low temperature scale and also suggests that the conduction electron density is low ($n_c \sim 0.5/atom$).

The samples were single crystals of YbAl$_3$ and LuAl$_3$ grown by the "self-flux" method in excess Al. The quality of such samples, as measured by the resistance ratio ($R(300K)/R(2K) \sim 60$) is sufficiently high that dHvA signals are well-resolved[9]. The susceptibility was measured using a SQUID magnetometer and the specific heat was measured via a relaxation technique. The Hall coefficient was measured in a magnetic field of 1T using an a.c. resistance bridge. The magnetoresistance was measured at the Los Alamos Pulsed Field Facility of the National High Magnetic Field Laboratory using a 20T superconducting magnet and an a.c. bridge. The $4f$ occupation number $n_f(T)$ was determined from the Yb L_3 x-ray absorption near-edge structure, measured at the Stanford Synchrotron Radiation Laboratory (SSRL) on beam line 4-3; the technique for extracting n_f from the data and other experimental details were similar to those discussed earlier [10]. We note here that the Lu L_3 near-edge structure measured for LuAl$_3$ was used a standard.

In Fig. 1 we plot the susceptibility $\chi(T)$ and the linear coefficient of the 4f specific heat, where $\gamma_m = C_m/T$ and $C_m = C(\text{YbAl}_3) - C(\text{LuAl}_3)$. (For LuAl$_3$ at low temperatures, $C = \gamma T + \beta T^2$ with $\gamma = 4$mJ/mol-K^2
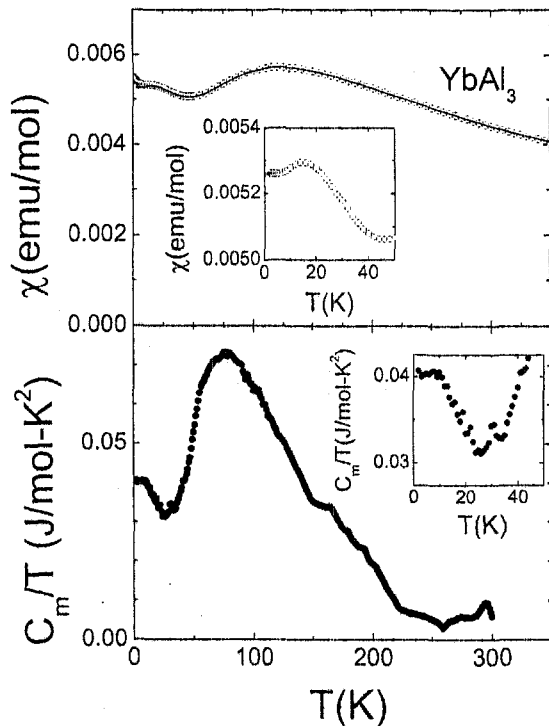
FIG. 1: (a) The magnetic susceptibility $\chi(T)$ and (b) the magnetic contribution to the specific heat coefficient C_m/T for YbAl$_3$. The insets exhibit the low temperature behavior; a small Curie tail has been subtracted from the data in the inset for $\chi(T)$.

and $\beta = 1.15 \times 10^{-4}$ J/mol-K^4, which implies a Debye temperature $\Theta_D = 257$K.) The broad peaks near 100K are typical of Yb IV compounds with $T_K \sim 500$K. In addition, there is a peak at 15K in the susceptibility (first reported by Heiss et al.[11]) and the specific heat coefficient displays an upturn below 25K which saturates at $T = 0$. These additional features are the basic evidence for the existence of a low temperature scale, $T_{coh} \lesssim 50$K below which there is a significant change in the behavior of the compound.

Evidence for this change of character can also be seen in the magnetotransport (Fig. 2). The Hall coefficient of LuAl$_3$ is typical of a metal, being constant and of a magnitude that in a one-band model implies a carrier density $n_c = 1/eR_H = 2e^-/f.u.$. The high temperature Hall coefficient of YbAl$_3$ varies with temperature in a manner suggestive of scattering from Yb moments (although the data cannot be fit well with the standard skew scattering formula[12]). Near 50K the derivative dR_H/dT of the Hall coefficient changes sign. Similar anomalies in the low temperature Hall coefficient have been observed in a number of Ce compounds and have been attributed to the onset of coherence[12]. The magnetoresistance (Fig. 2b,

inset) follows a B^2 law above 50K and the magnitude is approximately the same for field parallel and transverse to the current. Below 50K, the magnetoresistance becomes more nearly linear and the transverse magnetoresistance becomes substantially larger ($\Delta R/R \sim 0.75$) than the parallel magnetoresistance ($\Delta R/R \sim 0.35$) at 2K. In Fig. 2b we plot $\Delta R/R$ versus Br_0 where $r_0 = R(150K)/R(T)$; this tests Kohler's rule, i.e. that at any temperature $\Delta R/R = Af(Br_0)$ depends only on the product Br_0. The data violate this rule essentially because A varies with T, increasing by a factor of almost 1.5 between 40 and 80K; this crossover is seen most clearly in the data measured as a function of temperature at a fixed field 17.5T. These magnetotransport anomalies suggest that the anomalies in χ and C/T may be associated with an alteration of the Fermi surface. The fact that the resistivity follows a T^2 law below 30K[9] suggests that the anomalies are also connected with the full establishment of Fermi liquid coherence.

To demonstrate that the crossover from Fermi liquid behavior to local moment behavior is slower than predicted by the AIM we compare the experimental data to the AIM result that follows from the measured ground state values of the susceptibility and $4f$ occupation number[5, 7]. For fixed spin-orbit splitting ($\Delta_{so} = 1.3$eV), the theoretical results calculated in the non-crossing approximation (NCA) depend on three parameters: the f-level position E_f, the hybridization V between the $4f$ and the conduction electrons, and the width W of the conduction band, assumed Gaussian $N(E) = e^{-E^2/W^2}/\sqrt{\pi}W$. Since only states within $k_B T_K$ of the Fermi energy contribute significantly to the results, we argue that use of a Gaussian bandshape is acceptable as long as the density of states at the Fermi energy is chosen appropriately. We choose W to give the same specific heat coefficient that we observe in LuAl$_3$ ($\gamma = 4$mJ/mol-K^2). Once W is fixed, the values of E_f and V that yield the appropriate $n_f(0)$ and $\chi(0)$ can be determined uniquely. The values of the parameters are given in Fig.3; T_K is determined from the formula

$$T_K = \left(\frac{V^2}{\sqrt{\pi}W|E_f|}\right)^{1/8}\left(\frac{W}{\Delta_{so}}\right)^{6/8}We^{\sqrt{\pi}WE_f/8V^2}$$

which includes the effect of spin orbit splitting but ignores crystal field splitting since for IV compounds $T_K \gg T_{cf}$. It is clear from Fig. 3 that the susceptibility, $4f$ occupation number and $4f$ entropy $S_m = \int dT\, C_m/T$ all qualitatively follow the predictions of the AIM. The calculated coefficient of specific heat ($\gamma = 47.8$mJ/mol-K^2) is within 20% of the measured value ($\gamma_m = 40.65$mJ/mol-K^2). Indeed the data even are in accord with the prediction that the entropy $S_m(T)$ approaches the high temperature limit faster than the effective moment $T\chi/C_J$ which in turn evolves more rapidly than $n_f(T)$ (see Fig. 3). Nevertheless, it is also clear that the experimental data for these quantities approach the high temper-
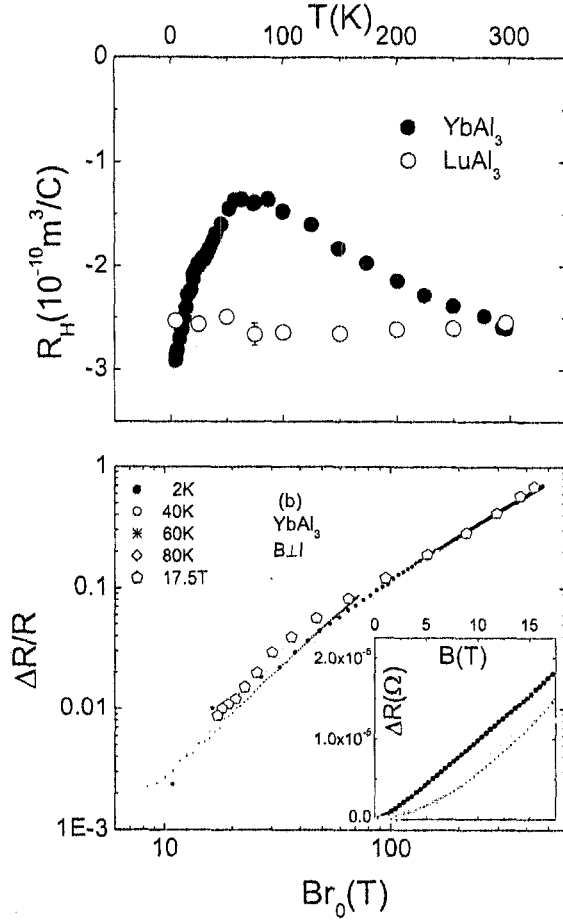
FIG. 2: a) The Hall coefficient of YbAl₃ (closed circles) and LuAl₃ (open circles) versus temperature. For LuAl₃ a typical error bar is shown; for YbAl₃ the error is smaller than the size of the symbols. b) Inset: The transverse magnetoresistance $\Delta R/R = R(H,T) - R(0,T)$ versus magnetic field for four temperatures. Main panel: The relative magnetoresistance $\Delta R/R$ versus Br_0 where $r_0 = R(150K/R(T))$.

ature limit considerably more slowly than predicted by the AIM theory.

Thus we have demonstrated the existence of a new low temperature scale for YbAl₃ and we have shown that the crossover to local moment behavior is slower than expected based on the Anderson Impurity Model. In the theory of the Anderson lattice, such results are expected[7, 8] as the conduction electron density decreases from the value $n_c = 1$ appropriate to a half-filled band when $n_f = 1$. In our earlier work[5] on YbXCu₄ we assumed that the background band in the Yb compound is the same as the measured band in the corresponding Lu compound. Using a one-band formula $n_c = 1/eR_H(LuXCu_4)$ we found that the slow crossover emerged when the number of electrons per atom (the number per formula unit divided by the number of atoms

in the formula unit) decreased below the value unity. Strong deviations occurred already for YbMgCu₄ where $n_c \sim 0.5$/atom. Using the same approximations for YbAl₃, we deduce from the Hall coefficient of LuAl₃ (Fig. 2a) that $n_c \sim 0.5$/atom. Hence, while the conduction electron density is not low for YbAl₃, it is (in this approximation) as low as in other compounds where strong deviations from the AIM are observed.

We believe that such effects are generic to IV compounds. A number of years ago we gave evidence[13] for a small coherence scale in the IV compound CePd₃ based on a low temperature peak in the susceptibility and the extreme sensitivity of the transport behavior to Kondo hole impurities below 50K. A low temperature anomaly in the Hall coefficient also has been observed for this compound[12]. Optical conductivity measurements[2, 14, 15] showed both that the temperature scale for these effects is the same as for the renormalization of the effective mass and the onset of the hybridization gap and that the conduction electron density of CePd₃ is less than 0.1 carrier/atom. Hence the anomalies appear to be associated both with the onset of the fully renormalized coherent ground state and with low conduction electron density. Measurement of the infrared optical conductivity is clearly a key future experiment for YbAl₃.

Neutron scattering measurements on polycrystals of YbAl₃[16] indicate that in addition to the usual Lorentzian excitation centered at $E_0 = 40$meV, corresponding to the high temperature Kondo scale (for the parameters of Fig. 3 the AIM predicts $E_0 = 39.6$meV), a new excitation centered at 30meV arises below 50K. Preliminary results[17] in single crystals also exhibit this excitation. However, the scattering does not exhibit the expected decrease with increasing $|Q = q + G|$ (where G is a reciprocal lattice vector) for Yb $4f$ scattering, which should follow the Yb 4f form factor. Indeed the scattering appears to be independent of G. Further work needs to address this issue.

The theoretical work on the Anderson lattice which predicts slow crossover and a low temperature coherence scale was motivated by the desire to understand "Noziéres' exhaustion". This concept was raised[18] to try to understand how the conduction electrons could screen the $4f$ spins when the number of conduction electrons n_c is smaller than the number n_f of $4f$'s in the lattice. The theoretical work to date has all been performed in the Kondo limit. We have examined the extension of the slave boson mean field theory for the Anderson lattice to the case $n_f < 1$ and $n_c < 1$ relevant to YbAl₃, where n_f is the number of holes in the $(2J + 1)$ fold degenerate f level. Following Millis and Lee[19], we define the Kondo temperature k_BT_K as the energy of the renormalized f level relative to the Fermi-energy. The coherence scale is defined as the renormalized (quasiparticle) $T = 0$ bandwidth which for a background conduction
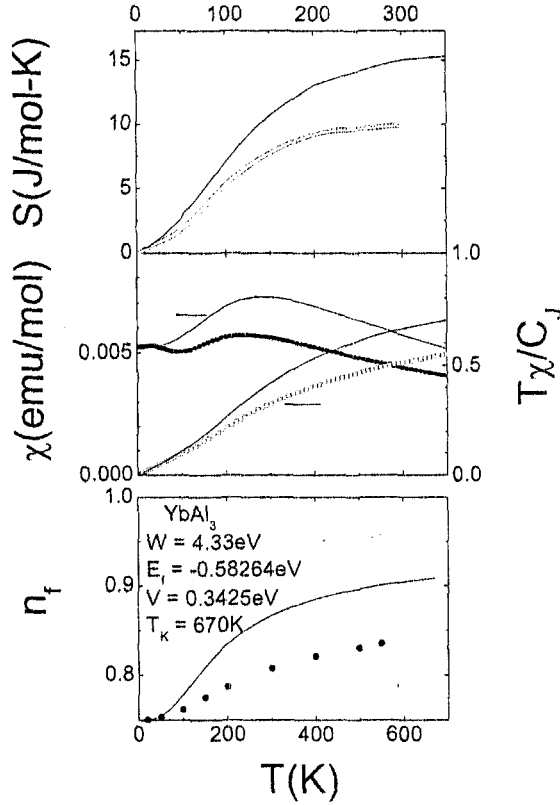
FIG. 3: (a) The $4f$ entropy S_m, (b) the susceptibility $\chi(T)$ (solid symbols) and the effective moment $T\chi/C_J$ (open symbols) where C_J is the $J = 7/2$ Curie constant and (c) the $4f$ occupation number $n_f(T)$ for YbAl$_3$. The symbols are the experimental data and the solid lines are the predictions of the Anderson Impurity Model (AIM), with input parameters given in the figure.

band density of states ρ is given by $k_B T_{coh} = \rho \tilde{V}^2$ where $\tilde{V}$ is the renormalized hybridization $\tilde{V} = \sqrt{1 - n_f} V$. In the limit $(\rho\tilde{V})^2 << n_c n_f/(2J + 1)^2$ we find that $T_{coh}/T_K = n_f/(2J + 1)$ independent of n_c. For YbAl$_3$ this means that T_{coh} should be an order of magnitude smaller than T_K, in qualitative agreement with the data.

In any case we assert that the two energy scales and slow crossover predicted by the theory are features of our data, that these effects show some correlation with a standard measure of the conduction electron density and that they may be generic for IV compounds.

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